Nano-crystalline Aluminium alloys based on Al-Fe, Al-Cu and Al-Sc systems

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Introduction

Recently, Sasaki et al [1] demonstrated that mechanically alloyed Al-5Fe alloy consolidated by spark plasma sintering (SPS) exhibited compressive yield strength of 1GPa and a large plastic strain of 30%. The high strength in this case was attributed to the nanocrystalline grain size of a-Al (~80nm) and fine distribution of Al_6Fe, while the ductility was attributed to larger aluminium grains. The Al-5Fe alloy developed by Sasaki et al [1] also retained significantly high compressive yield strengths at elevated temperatures with compressive yield strength of ~500MPa measured at 350^{o}C. The elevated temperature properties of this alloy are higher than that reported for other nano-crystalline Al-TM based alloys. In order to enhance the ductility of Al-5Fe alloy the high volume fraction of Al_6Fe and $Al_{13}Fe_4$ phases must be reduced but without any cost to the nanocrystalline grain size observed in order to preserve high yield strengths observed. The prior investigations of Al-Fe-Zr system [2,-4] and use of Zr as the grain refiner in the conventional aluminum alloys [5] suggest that addition of small amount of Zr to leaner Al-Fe alloy may lead to development of a high strength nanocrystalline Al-Fe-Zr alloy processed via SPS.

In this report the effect of Zr additions on the SPS processed mechanically alloyed Al-1Fe alloys on the microstructure, mechanical properties will be presented. Additional work on the effect of other solute additions such as Cu and Sc on nano-crystalline Al alloys will also be presented.

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14. ABSTRACT
Bulk nano-crystalline Al-1Fe-0.5Zr alloys were fabricated via mechanical alloying and spark plasma sintering processing exhibited high compressive yield strength of 854MPa with a plastic strain of 19% at room temperature. Compressive yield strength of 455MPa was exhibited at 250oC. A high strength and plastic strain was exhibited by these alloys with substantially lower amount of solute used as compared with other alloys with similar strengths. The high strength and high plastic strains has been attributed to the presence of high density of nano-crystalline grains with grain size of 65nm and a small volume faction of coarse grains distributed through the microstructure. Control of mechanical alloying process and SPS condition can be utilized to tailor the mechanical properties of nano-crystalline Al alloys through control of the microstructure.

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Experimental procedure

Pure Al (99.9% purity and 53-106mm diameter), Fe (99.9% purity and 53-106mm diameter), Cu (99.9% purity and 53-106mm diameter) and Zr (99.9% purity and 53-106mm diameter) powders and Al_3Zr and Sc_2O_3 powders were used in the present investigation. Al_3Zr powders were prepared by arc melting pure Al and Zr at stochiometric composition of Al_3Zr and crushing the alloy ingot produced. Mechanical alloying was carried out at ambient temperature using a Fretsch Pulverisette P-6 planetary ball mill in a hardened stainless steel container using 10mm diameter balls made of same material. The powder to ball ratio was kept at 1:10 with 6wt% Ethanol as the process control agent with a milling speed of 250rpm with milling time of 90h.

The mechanically milled powders were consolidated using SPS machine, Sumitomo Coal Mining Company model 1050. In present work a tungsten carbide die with an inner diameter of 10mm used to consolidate ~1.5g of powder. The powders were sintered under a vacuum of ~2×10^{-3}Pa under a 35kN load at a 420 or 480^{o}C for 10min. Samples were heated at 2^{o}C/s

Alloyed powders and sintered samples were characterized via X-ray diffraction (XRD) analysis on a RIGAKU RINT2500 X-ray diffractometer with Cu K_a radiation to estimate the grain size and the amount of Fe and Zr dissolved in Al in the milled powders and to determine the constituent phases in the sintered samples. Microstructures of sintered samples were characterized with scanning electron microscopy (SEM), using Carl Zeiss 1540 Energy Selective Backscattered instrument operating at 2kV and 15kV and transmission electron microscopy (TEM) using Tecnai G^2 F30 instrument operating at 300kV. Thin foil specimens for TEM were prepared by punching 3 mm diameter discs and twin jet electro-polishing in a solution of methanol and nitric acid with a ratio of 2:1 at ~-50^{o}C at a voltage of 20V.

Al-Fe-Zr system

As milled powder

The as milled powders were characterised with the use of XRD and showed single phase aluminium in the as milled condition for up to 1at% Zr, Figure 1 (a). This is significantly higher than the equilibrium solid solubility of Zr in Al which is ~0.1at%. The dissolution of up to 3at%Zr has been achieved previously through mechanical milling for approximately 90h using Al_3Zr powders as precursor for Zr additions. The as milled grain size decreased with the

increased Zr content up to 0.5at%Zr and increased with further additions, Figure 1 (b). The grain size was minimised at a Zr concentration of 0.5at%.

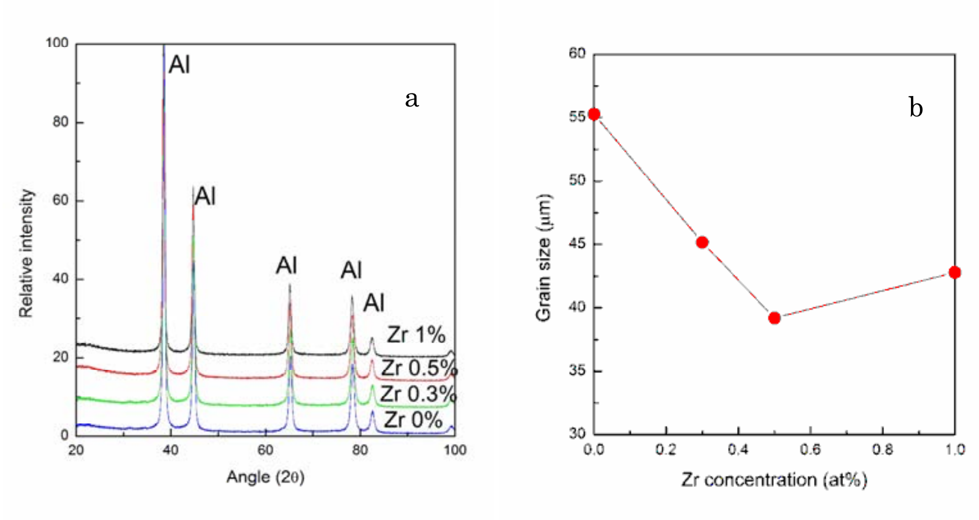


Figure 1 (a) XRD spectra showing complete dissolution of Zr in the Al-1Fe alloy during mechanical milling for 90h and (b) grain size variation with the Zr additions following mechanical milling.

Compressive behaviour at room temperature

Following SPS processing at 480°C the compressive behaviour of Al-1Fe-(0-1at%) and the compressive yield strength was measured and the maximum yield strength was achieved for 0.5at% alloy, Figure 2 (a). The SPS temperature was then optimised in the temperature region 400-480°C, Figure 2 (b). The optimised SPS condition for the Al-1Fe-0.5Zr alloy was achieved at 420°C after mechanical alloying for 90h. The compressive yield strength and the plastic strain at failure observed for the Al-1Fe sample was significantly less than the yield strength reported by Sasaki et al [1] using Al-5Fe samples but showed similar plastic strains at failure. The addition of Zr enhanced the compressive yield strength observed but the optimised yield strength achieved; SPS processing at 420°C was somewhat lower than that observed for the Al-5Fe alloy at 854MPa.

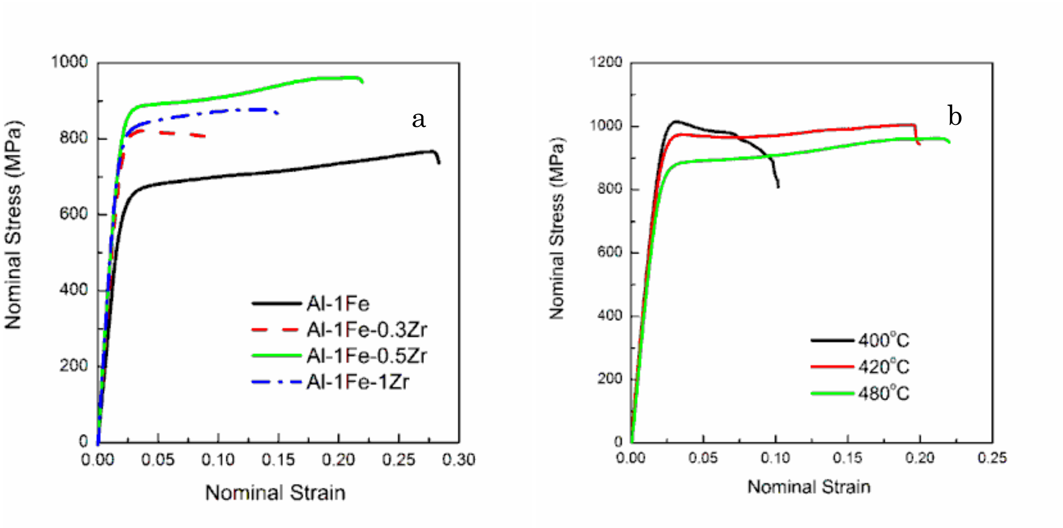


Figure 2 The compressive stress strain behaviour of (a) samples of Al-1Fe-(0-1)Zr alloys SPS processed at 480°C and (b) samples of Al-1Fe-0.5Zr alloys with varying SPS temperature.

Elevated temperature behaviour

The elevated temperature compression properties were measured for the Al-1Fe-0.5Zr alloy mechanically milled for 90h and sintered at 420°C. The variation of 0.2% yield strength with test temperature is shown in Figure 3. The data is plotted alongside data from a previous study on the elevated temperature properties of SPS processed Al-5Fe [1] was investigated. This showed a continued decrease in the compressive yield strength with increased testing temperature however there was a sharp decrease with the increase of test temperature to 350°C. The elevated temperature properties exhibited by the Al-1Fe alloy were comparable with the Al-5Fe alloy in the temperature range150-250°C. With the increase in testing temperature to 350°C the yield strength of Al-1Fe-0.5Zr alloy deteriorated.

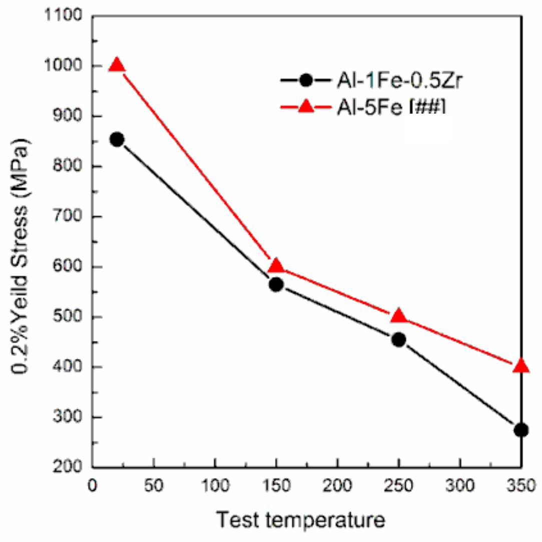


Figure 3 Variation of yield strength with increase in the testing temperature for Al-1Fe-0.5Zr alloy, Al-5Fe alloy from [1].

Microstructures

The microstructures of the SPS processed Al-1Fe-xZr (0-1) (at%) samples spark plasma sintered (SPS) at 480°C were examined with the aid of SEM, Figure 4(a-d). The back scattered SEM images showed both bright and dark contrast. In this case the bright contrast phase is mainly Al_6Fe with some contributions from Al_3Zr phase. The dark contrast phase is Al rich Al-Fe solid solution. In the microstructure intermittently a phase that images black has been observed and it is likely this is pure Al. The Al-1Fe sample showed in addition to a coarse distribution of intermetallic particles, some coarse grains of pure Al grains were also observed, Fig 4 (a). Addition of Zr refined the distribution of intermetallic particles but with 0.3at%Fe there are many large intermetallic particles present within the microstructure. Increase in the Zr content to 0.5at%r refined the precipitate distribution significantly. However with further addition of Zr to 1at% the particle size coarsened. The reduction of SPS temperature to 420°C for the Al-1Fe-0.5Zr alloy, Fig 4 (e), resulted in a fine and uniform distribution of intermetallic particles through the microstructure similar to sample processed at 480°C

In order to understand the microstructural differences between the Al-1Fe-0.5Zr sample treated at 480°C and 420°C microstructures were investigated with the use of TEM, Figure 5. In both instances, the microstructure consisted of regions of coarse grained Al and nano-crystalline grains of Al. Coarse grained Al found in the samples processed at 480°C was slightly larger than that formed when processed at 420°C, Fig 5 (a-b). The nano crystalline regions observed in the both samples are shown in Fig 5 (c-f) and expected to be randomly oriented given the ring patterns recorded for the electron diffraction work. Using the dark field images of nano crystalline region the grain size was elucidated and a grain size of approximately 83 and 65nm observed for the samples processed at 480 and 420°C, respectively.

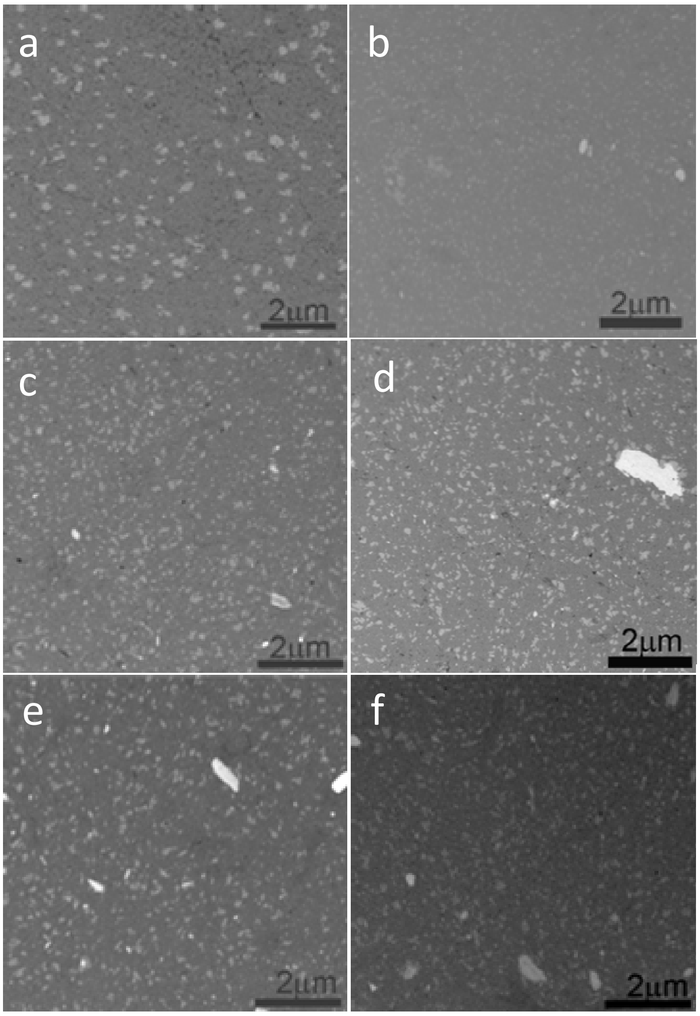


Figure 4 SEM microstructure typical of (a-d) Al-1Fe-(0-1)Zr samples processed at 480°C. Al-1Fe-0.5Zr alloy processed at (e) 420°C and (f) 400°C.

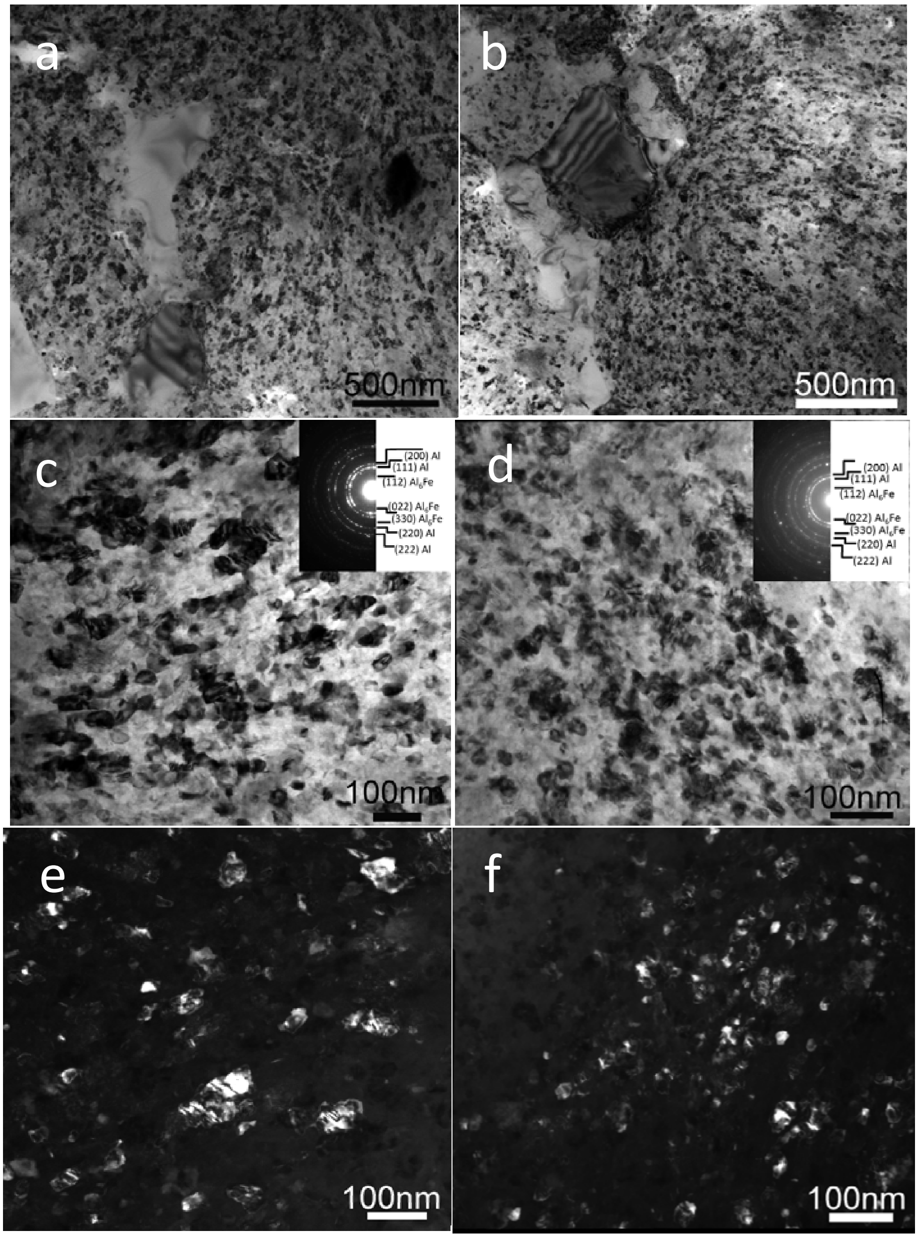


Figure 5 TEM microstructures of Al-1Fe-0.5Zr alloy SPS processed at (a ,c, and e) 480^{o}C and (b, d, and f) 420^{o}C. (a,b) Coarse grained region (c-f) nano-scaled grained region, (c,d) are bright field images and (e, f) are dark field images recorded using part of <111>$_{Al}$ ring.

Effect of increasing Fe content

In order to enhance the yield strength of Al-1Fe-0.5Zr alloy from the optimised value of 854MPa the Fe content was further increased to 2at%. The increase in the Fe content did not result in the repulsion of Zr from solid solution in Al, Figure 6 but the compression testing following SPS processing at 420^{o}C did not show any appreciable elongation at failure while higher yield strength of ~950MPa was observed. Therefore further work is necessary to optimise the processing conditions for the Al-2Fe-0.5Zr alloy.

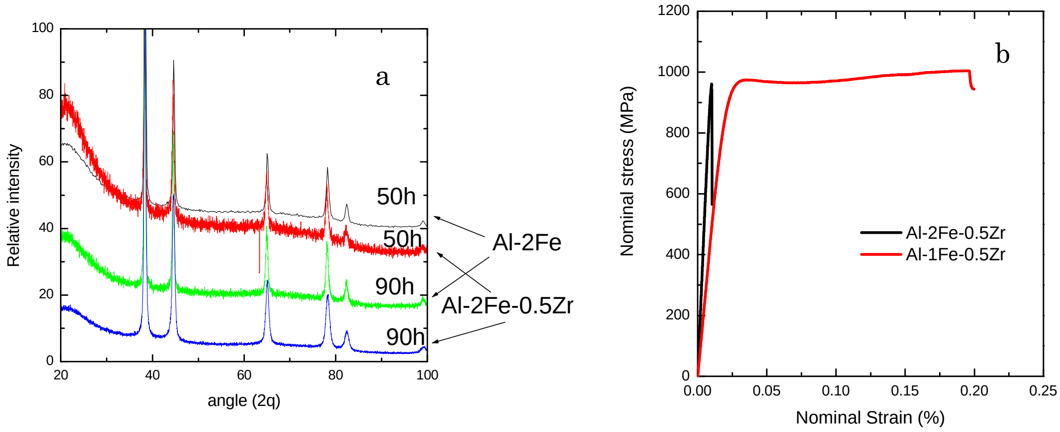
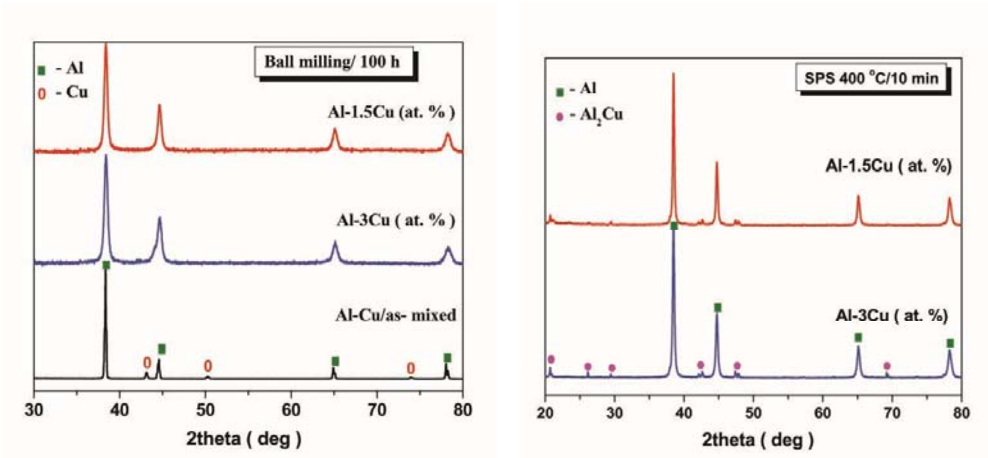


Figure 6 (a) XRD spectra from Al-2Fe and Al-2Fe-0.5Zr alloys mechanically milled for 50 and 90h and (b) compression properties of Al-2Fe-0.5Zr compared to Al-1Fe-0.5Zr both SPS processed at 420^{o}C for 10min.

Al-Cu and Al-Sc systems

It has been reported that the transition metal additions to Al results in enhancement of mechanical properties following mechanical milling of nano-crystalline Al. Therefore Cu was chosen at it has an appreciable solid solubility in Al. Following mechanical milling for 100h at ambient temperatures a complete solid solution of Cu in Al was achieved for up to 3at% Cu, Figure 7 (a) the supersaturated Al solid solution decomposed into Al and Al_2Cu following SPS processing at 400^{o}C for 10min, Figure 7 (b). The mechanical properties of the SPS processed alloys showed compressive yield strength of approximately 700MPa and 820 were achieved for the 1.5 and 3at% Cu containing samples respectively, Figure 8. The microstructures of Al-1.5Cu alloy showed a distribution of very fine scale Al_2Cu particles dispersed through the microstructure providing the appropriate strengthening Figure 9.



a b

Figure 7 XRD spectra recorded from Al-(1.5 -3) Cu alloys in the (a) as mechanically alloyed condition (b) following SPS processing at 400^{o}C for 10min.

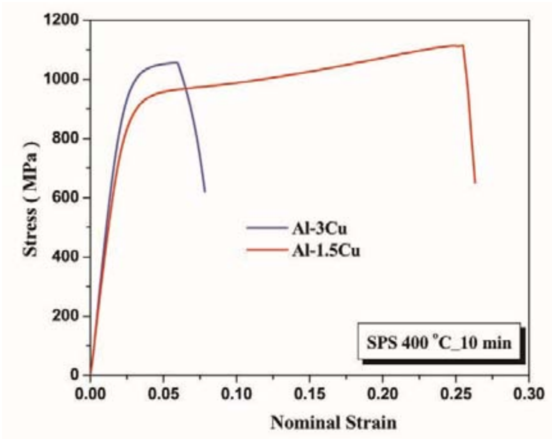


Figure 8 Compression properties of Al-1.5Cu and Al-3Cu following SPS processing

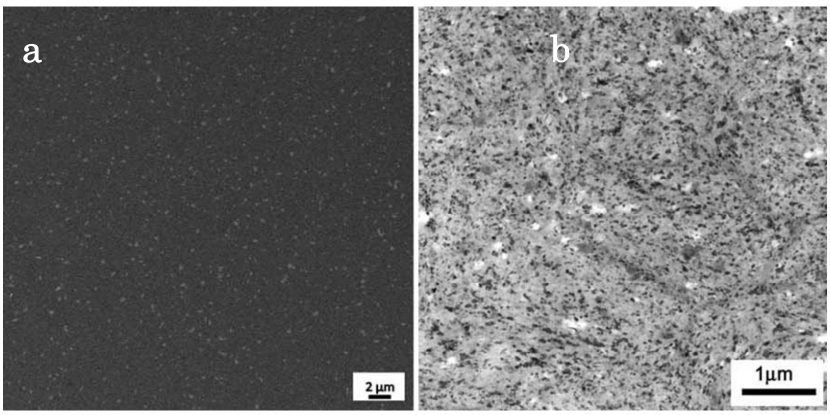
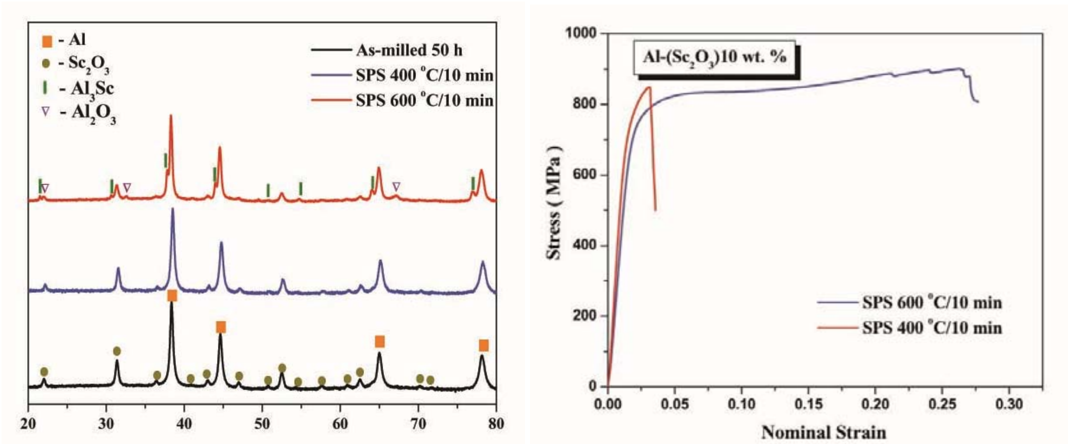


Figure 9 microstructures of Al-1.5Cu alloy (a) SEM and (b) TEM

The Sc was added to the Al powders as Sc_2O_3 which remained as is during mechanical milling but decomposed to form Al_2Sc during SPS processing at both 400 and 600°C, Figure 10 (a). The resultant compression properties showed that processing at 600°C enhanced the ductility while the yield strength reduced to 650MPa from the 730MPa observed for 400°C, Figure 10 (b). The resultant ductility has been attributed to the increased size of Al_2Sc particles and coarse distribution of Al grains through the microstructure, compared with samples SPS processed at 400°C, Figure 11.



a b

Figure 10 (a) XRD spectra recorded from Al10wt%Sc_2O_3 containing alloy in the as mechanically milled and SPS processed conditions. (b) Compression properties of SPS processed alloys

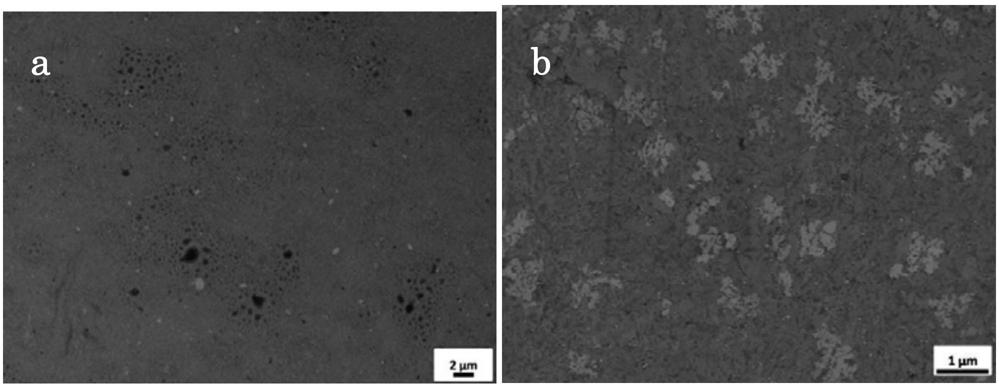


Figure 11 SEM microstructures of SPS processed alloys (a) 400°C for 10min and (b) 600°C for 10min.

Conclusions

Bulk nano-crystalline Al-1Fe-0.5Zr alloys fabricated via mechanical alloying and SPS processing exhibited high compressive yield strength of 854MPa with a plastic strain of 19% at room temperature. Compressive yield strength of 455MPa was exhibited at 250^{o}C. A high strength and plastic strain was exhibited by these alloys with substantially lower amount of solute used as compared with other alloys with similar strengths. The high strength and high plastic strains has been attributed to the presence of high density of nano-crystalline grains with grain size of 65nm and a small volume faction of coarse grains distributed through the microstructure. Control of mechanical alloying process and SPS condition can be utilised to tailor the mechanical properties of nano-crystalline Al alloys through control of the microstructure.

References

[1] T.T. Sasaki, T. Ohkubo and K. Hono Acta Mater. 3529-3538

[2] H. Sasaki, N. Kobayashi, K, Kita J. Nagahora A. Inoue Mater. Trans. 44 (2003) 1948-195

[3] C.A.D. Rodrigues D.R. Leiva K.R. Cardoso, C.S. Kiminami W.J. Botta Mater. Sci. Forum 386-388 (2002) 33-38

[4] G. Vigier U. Oritiz Mendez, P. Merele, G. Thollet F. Fougut Mater. Sci. Eng 98 (1988) 191-195

[5] I.J. Polmear Light alloys 4th edition Butterworth Heinemann (2006) Oxford.